

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau



(43) International Publication Date
16 January 2003 (16.01.2003)

PCT

(10) International Publication Number
WO 03/005465 A1

(51) International Patent Classification⁷: **H01M 4/40**, 4/52

(21) International Application Number: PCT/US01/43820

(22) International Filing Date:

19 November 2001 (19.11.2001)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:

PCT/US01/13308 25 April 2001 (25.04.2001) US

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(81) Designated States (national): AE, AG, AL, AU, BA, BB, BG, BR, BZ, CA, CN, CO, CR, CU, CZ, DM, DZ, EC, EE, GD, GE, HR, HU, ID, IL, IN, IS, JP, KP, KR, LC, LK, LR, LT, LV, MA, MG, MK, MN, MX, MZ, NO, NZ, PL, RO, SG, SI, SK, TR, TT, TZ, UA, US, UZ, VN, YU, ZA.

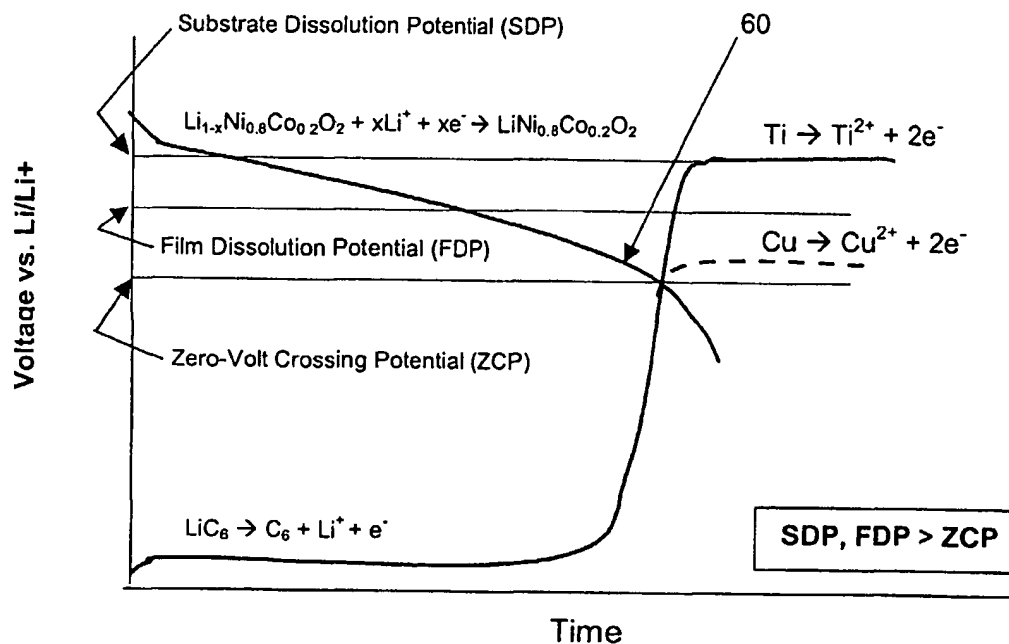
(84) Designated States (regional): ARIPO patent (GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE, TR), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

Published:

— with international search report

[Continued on next page]

(54) Title: RECHARGEABLE LITHIUM BATTERY FOR TOLERATING DISCHARGE TO ZERO VOLTS



(57) Abstract: A lithium ion battery particularly configured to be able to discharge to a very low voltage, e.g. zero volts, without causing permanent damage to the battery. More particularly, the battery is configured to define a Zero Volt Crossing Potential (ZCP) which is lower than a Damage Potential Threshold (DPT).



For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

**TITLE : RECHARGEABLE LITHIUM BATTERY FOR TOLERATING
DISCHARGE TO ZERO VOLTS**

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FIELD OF THE INVENTION

This invention relates generally to rechargeable electric batteries particularly suited for applications, e.g., implanted medical devices, where a battery cannot be easily replaced. More particularly, the invention relates to rechargeable lithium batteries configured to tolerate deep discharging to zero
10 volts without permanently damaging the battery's energy storing capacity.

BACKGROUND OF THE INVENTION

Rechargeable electric batteries are employed in a wide range of applications, e.g., consumer products, medical devices, and aerospace / military
15 systems, which respectively impose different performance requirements. In some applications, e.g., implanted medical devices, it is important that the battery be able to reliably maintain its performance characteristics over a long useful life despite extended periods of inactivity. Implanted medical device applications impose special requirements on a battery because the medical
20 device needs to be highly reliable to perform critical tasks, the battery may remain inactive and uncharged for extended periods, e.g., several months, and it is difficult and/or expensive to replace a battery. Analogous conditions exist in various aerospace / military applications. For example, a rechargeable battery may be deployed to power a satellite in deep space where it cannot be
25 replaced and must be able to operate over a long life under varying conditions, including long periods of inactivity. Military applications often demand similar performance specifications since military hardware can be unused for several months but must remain ready to be activated. Current battery technology requires stored batteries to be charged every few months to avoid a permanent
30 reduction in energy storing capacity.

In order to avoid unnecessary surgery to replace a damaged battery in an implanted medical device, it is desirable that a battery perform reliably over a very long life , i.e., several years ,under a variety of conditions. Such

conditions can include extended periods of non-use which may allow the battery to deeply self discharge to zero volts. It is typical for prior art rechargeable lithium batteries to suffer a permanent capacity loss after discharging below 2.5 volts. To avoid such capacity loss, it is important to regularly charge prior art
5 lithium batteries.

Existing rechargeable lithium batteries typically consist of a case containing a positive electrode and a negative electrode spaced by a separator, an electrolyte, and feedthrough pins respectively connected to the electrodes and extending externally of the case. Each electrode is typically formed of a
10 metal substrate that is coated with a mixture of an active material, a binder, and a solvent. In a typical battery design, the electrodes comprise sheets which are rolled together, separated by separator sheets, and then placed in a prismatic or cylindrical case. Positive and/or negative feed through pins (i.e., terminals) are then connected to the respective electrodes and the case is filled with
15 electrolyte and then sealed. The negative electrode is typically formed of a copper substrate carrying graphite as the active material. The positive electrode is typically formed of an aluminum substrate carrying lithium cobalt dioxide as the active material. The electrolyte is most commonly a 1:1 mixture of EC:DEC in a 1.0 M salt of LiPF_6 . The separator is frequently a microporous membrane
20 made of a polyolephine, such as a combination of polyethylene and/or polypropylene which can, for example, be approximately 25 microns thick.

Batteries used in implanted medical devices can be charged from an external power source utilizing a primary coil to transfer power through a patient's skin to a secondary coil associated with the implanted medical device.
25 The secondary coil and an associated charging circuit provide a charging current to the battery. Protection circuitry is typically used in conjunction with prior art lithium batteries to avoid the potential deleterious effects of over or under charging the battery. Such protection circuitry can terminate charging if the voltage or temperature of the battery exceeds a certain level. Moreover, it
30 is common to also incorporate low voltage protection to disconnect the battery from its load if the voltage of the battery falls below a certain lower level. This latter precaution is taken to prevent permanent damage to the battery that will

likely occur if the voltage on an electrode exceeds a Damage Potential Threshold (DPT). For example, it is well known in the industry that discharging a lithium battery to below 2.5 volts and storing it for an extended period of time will likely result in a permanent loss of battery capacity. Despite incorporating
5 low voltage cutoff protection to disconnect the battery from its load if the voltage falls below a certain threshold, typical prior art batteries will slowly self-discharge further causing the voltage of an electrode to exceed the Damage Potential Threshold.

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SUMMARY OF THE INVENTION

The present invention is directed to a rechargeable lithium battery particularly configured to permit it to discharge to a very low voltage, e.g. zero volts, without causing permanent damage to the battery. More particularly, a battery in accordance with the invention is configured to define a Zero Volt
15 Crossing Potential (ZCP) which is lower than the battery's Damage Potential Threshold (DPT).

ZCP refers to the voltage on the positive and negative electrodes relative to a lithium reference (Li/Li+) when the battery potential, i.e., the voltage between the electrodes, is zero. The Damage Potential Threshold (DPT) is
20 attributable to at least two factors, i.e., a Substrate Dissolution Potential (SDP) and a Film Dissolution Potential (FDP). SDP refers to the voltage of the negative electrode, relative to the lithium reference, above which the electrode substrate starts to corrode or decompose to permanently damage the substrate. FDP refers to the voltage of the negative electrode, relative to the lithium
25 reference, above which a solid electrolyte interface (SEI) layer begins to dissolve. The SEI, or film, comprises a passivation layer which, in normal operation, forms on the negative electrode and functions to inhibit a continuing reaction between the negative electrode active material and the electrolyte. When the voltage of the negative electrode relative to the lithium reference,
30 exceeds either SDP or FDP, physical damage to the electrode is likely to occur thereby permanently impairing the battery's capacity.

A battery's ZCP level relative to the lithium reference is dependent in part on the materials used for the positive and/or negative electrodes. In accordance with a preferred embodiment of the invention, a positive electrode active material $\text{LiNi}_x\text{Co}_{1-x}\text{O}_2$ is selected which exhibits a discharge voltage curve
5 appropriate to achieve a relatively low Zero Crossing Potential (ZCP) level. This feature of the preferred embodiment facilitates the implementation of a battery characterized by a ZCP less than its Damage Potential Threshold (DPT). It has been recognized that as more Ni is substituted for Co (i.e., increasing x), the magnitude of the discharge voltage profile decreases. It has been determined
10 that values of x between 0.5 and 1.0 optimally achieve the desired ZCP/DPT relationship in accordance with the present invention, i.e., $\text{LiNi}_x\text{Co}_{1-x}\text{O}_2$ (where $0.5 \leq x \leq 1.0$). For $x < 0.5$, there is a noticeable degradation in capacity retention after storage at zero volts indicating that some corrosion or internal battery degradation has occurred at low voltage similar to the results seen from a
15 conventional lithium battery design.

In accordance with a preferred embodiment of the invention, $\text{LiNi}_{0.8}\text{Co}_{0.2}\text{O}_2$ is used for the positive active material on a thin metal substrate, e.g., aluminum. The negative electrode is preferably formed of a titanium or titanium alloy substrate carrying a layer of negative active material, e.g., graphite coated on
20 both faces of the substrate.

Batteries in accordance with the present invention are particularly suited for use in critical applications where physical access to the battery is difficult and/or expensive, such as in medical devices configured to be implanted in a patient's body. Such a medical device is typically comprised of a hermetically
25 sealed housing formed of biocompatible material and dimensioned to be implanted without interfering with normal body function. A battery in accordance with the invention includes a case configured for mounting in the device housing. The battery case can be of a variety of shapes, e.g., prismatic or cylindrical, and typically defines a volume of between .05 cc and 30 cc.
30 Batteries within this range exhibit capacities between 1.0 milliamp hours and 10 amp hours. An exemplary battery for use in a neurostimulation device includes a prismatic hermetically sealed battery casing having dimensions of 35 mm x

17mm x 5.5 mm. A wide variety of medical device applications are discussed in the medical and patent literature; see, for example, US Patent 6,185,452.

BRIEF DESCRIPTION OF THE DRAWINGS

5 The above and other features and uniqueness of the invention will be better visualized from the following drawings and schematics.

Figure 1A schematically depicts positive and negative battery electrodes rolled around a mandrel for placement in a battery case and Figure 1B depicts in cross-section a complete battery;

10 Figure 2 shows a typical deep discharge curve for a conventional lithium ion battery using copper as the negative electrode substrate and lithium cobalt dioxide LiCoO_2 as the positive electrode active material;

Figure 3 shows a typical deep discharge curve for a lithium ion battery in accordance with the present invention using titanium as the negative electrode
15 substrate;

Figure 4 shows a typical deep discharge curve for a lithium ion battery in accordance with the present invention using $\text{LiNi}_x\text{Co}_{1-x}\text{O}_2$ ($0.5 \leq x \leq 1.0$) as the positive electrode active material;

Figure 5 is a table showing test results of various battery configurations
20 including a preferred embodiment in accordance with the present invention; and

Figure 6 schematically depicts a battery in accordance with the invention contained within an implantable medical device housing.

DETAILED DESCRIPTION

25 A rechargeable battery in accordance with the present invention is particularly suited for use in medical devices intended to be implanted in a patient's body. Such medical devices are extensively discussed in the medical and patent literature. For example, US Patent 6,189,452 describes a Battery-Powered Patient Implantable Device utilizing a rechargeable battery depicted
30 in alternative constructions in Figures 8A through 8G. The present invention is directed to an improved rechargeable lithium battery, useful in devices of the type described in Patent 6,185,452, configured to tolerate deep discharging

without significantly impairing the battery's ability to recover its original storage capacity.

Figures 1A and 1B schematically depict a typical lithium battery construction 10 comprising a prismatic case 12 containing a positive electrode 14 and a negative electrode 16, rolled around a mandrel 18. Separator sheets 20, 22 are incorporated in the rolling to electrically separate the electrodes. The case 12 also typically includes electrolyte material (not shown) and positive and negative feed through pins (i.e., terminals) 26, 28 which are respectively connected to the electrodes 14, 16 and extend externally of the case 12.

Typical prior art lithium ion batteries include a positive electrode 14 comprised of a thin metal substrate, e.g., aluminum, carrying a layer of positive active material, e.g., lithium cobalt dioxide LiCoO_2 mixed with a binder, and coated on both faces of the substrate. The negative electrode 16 is typically comprised of a thin metal substrate, e.g., copper, carrying a layer of negative active material, e.g., graphite coated on both faces of the substrate.

Two layers of separator 20, 22 electrically separate the electrodes 14, 16 from each other, enabling the electrodes to be rolled around mandrel 18. Each separator layer can comprise a micro porous membrane made of a combination of polypropylene and is approximately $25\mu\text{m}$ thick. The electrolyte is most commonly a 1:1 mixture of EC:DEC in a 1.0 M salt of LiPF_6 .

Figure 2 shows typical deep discharge performance curves for a conventional lithium ion battery. The y-axis represents voltage relative to a lithium reference (Li/Li^+) or counter electrode and the x-axis represents time. Curves 50 and 52 respectively depict the discharge curves for the positive and negative electrodes. The battery output voltage is the difference between the positive electrode voltage and the negative electrode voltage. During discharge, the positive electrode voltage decreases relative to the lithium reference and the negative voltage increases, primarily near the end of discharge. A protection or management circuit (not shown) is typically provided to disconnect the load to stop the discharge when the battery voltage reaches 2.5 volts. If, however, the discharge continues (attributable, for example, to self-discharge over a long period of time), the negative electrode potential will rise until it reaches the

potential of the positive electrode. This constitutes the Zero Volt Crossing Potential (ZCP) and is typically about 3.6 volts in conventional lithium ion battery constructions. The negative electrode potential at ZCP can exceed the Substrate Dissolution Potential (SDP) of the negative electrode substrate, e.g., 3.3 volts for copper, and cause decomposition and permanent damage to the substrate. The present invention is directed to battery improvements to assure that the value of SDP is greater than the value of ZCP, as represented in Figure 3.

Figure 3 depicts deep discharge performance curves for a lithium battery in accordance with the present invention in which the negative electrode substrate is formed of titanium instead of copper. The use of titanium increases the knee of the negative electrode curve 54 to position the SDP level above the ZCP level. This relationship considerably reduces potential damage to the negative electrode substrate. In addition to commercially pure titanium, i.e., titanium CP, other materials can be used to raise the SDP sufficiently, e.g., titanium alloys, nickel, nickel alloys, and stainless steel.

Figure 3 demonstrates how the SDP level can be increased relative to the ZCP by proper choice of the negative electrode substrate material. Alternatively, or additionally, the ZCP level can be decreased relative to the SDP by proper choice of the positive electrode active material, as depicted in Figure 4.

More particularly, Figure 4 shows the discharge curve 60 for a positive electrode using lithium nickel cobalt dioxide $\text{LiNi}_x\text{Co}_{1-x}\text{O}_2$ (where $0.5 \leq x \leq 1.0$) as the active material, i.e., as the intercalation compound. Note that the curve 60 of Figure 4 exhibits a greater negative slope than the analogous curve 50 of Figure 2 representing the standard intercalation compound LiCoO_2 . The effect of the increased negative slope is to lower the ZCP level relative to the lithium reference and the SDP level. As was the case in connection with Figure 3, this reduces the potential damage to the negative electrode substrate. Additionally, however, the ZCP level also falls below a Film Dissolution Potential (FDP) which is the voltage above which a solid electrolyte interface (SEI) layer begins to dissolve. The SEI, or film, comprises a passivation layer which forms on the

negative electrode and functions to inhibit a continuing reaction between the negative electrode active material and the electrolyte. Dissolution of the SEI can noticeably damage the negative electrode active material.

Experiments have been performed at two different temperatures
5 employing the aforescribed techniques depicted in Figures 3 and 4. The preliminary results are summarized in the table of Figure 5. Four different battery configurations were constructed as shown. Configuration (1) corresponds to the conventional arrangement represented in Figure 2 comprising a copper substrate for the negative electrode and LiCoO_2 for the
10 positive active material. The battery was built and then recycled once to get an initial capacity measurement. The battery was then shorted between the positive and negative leads to achieve a zero volt state. This zero volt condition was held for one week and then recharged and discharged to get a capacity measurement after zero-volt storage. The capacity retention is calculated by
15 dividing the discharge capacity after zero volt storage by the initial capacity and multiplying by 100%. In this manner, this percentage reflects any damage that had occurred to the battery while in the zero volt state.

As represented in Figure 5, the capacity retention for battery configuration (1) is below 80%, thus suggesting that the zero volt condition had
20 significantly damaged the battery. After opening the battery and examining the electrodes, it was seen that dissolution of the negative electrode copper substrate had occurred. This battery (1) configuration performed poorly at both temperature settings.

The battery configuration (2) used LiCoO_2 as the positive active material
25 and a titanium substrate as the negative substrate corresponding to the arrangement represented in Figure 3. The results show that at 25°C the capacity retention was at about 98% after the zero volt condition. However, at a higher temperature (37°C) exemplary of medical implant conditions, performance deteriorates to below 80%. This suggests that perhaps the zero
30 volt crossing potential was sufficiently below SDP to avoid substrate dissolution but still high enough to exceed FDP and cause damage to the negative electrode active material. Accordingly, attempts were made to lower ZCP further

to avoid damage both to the negative active material and the negative electrode substrate.

The battery configuration (3) utilizes $\text{LiNi}_x\text{Co}_{1-x}\text{O}_2$ (where $x=0.8$) as the positive electrode active material and a conventional copper negative electrode substrate. The results show that at 37°C , the capacity retention is quite high at 90%. However, examination after the test, revealed that some dissolution of the copper substrate had occurred. Battery configuration (4) uses both $\text{LiNi}_{0.8}\text{Co}_{0.2}\text{O}_2$ as the positive active material and titanium as the negative electrode substrate material. Results show that this configuration gives the best capacity retention after zero volt storage.

From the curves of Figure 4 and the table of Figure 5, it appears that some performance gain is achieved by configuration (2) using a titanium negative electrode substrate and by configuration (3) using $\text{LiNi}_x\text{Co}_{1-x}\text{O}_2$ (where $x=0.8$) as the positive active material. However, maximum performance gain appears in configuration (4) which combines both of these features.

Figure 6 schematically depicts a battery 60 in accordance with the invention mounted in a housing 64 (shown partially open for the purposes of illustration) of a medical device 66 configured for implanting in a patient's body. The housing 64 is preferably formed of biocompatible material and hermetically sealed. The device 66 is typically used for monitoring and/or affecting body parameters. For example, the device can be used to electrically stimulate nerves. The casing 68 of battery 64 can, for example, have dimensions of 35 mm x 17 mm x 5.5 mm. Other configurations and sizes are suggested in the literature, e.g., US Patent 6,185,452

While the invention has been described with reference to specific exemplary embodiments and applications, it should be recognized that numerous modifications and variations will occur to those skilled in the art without departing from the spirit and scope of the invention set forth in the appended claims.

CLAIMS

1. A rechargeable lithium battery configured to tolerate long periods of inactivity and keep discharging to zero volts, without prematurely reducing the battery's capacity, said battery comprising:
- a case containing a positive electrode and a negative electrode;
 - said positive electrode comprising a metal substrate having a lithium based active material formed thereon;
 - said negative electrode comprising a metal substrate having a carbon based active material formed thereon;
 - said positive and negative electrodes each defining a certain Zero Volt Crossing Potential (ZCP) relative to a lithium reference level when the voltage between said electrodes is zero;
 - said negative electrode substrate being susceptible of permanent damage when its voltage potential relative to said reference level exceeds a Damage Potential Threshold (DPT); and wherein
 - said positive and negative electrodes are formed of materials selected to establish ZCP at a lower level than DPT.
2. The battery of claim 1 wherein said positive electrode active material comprises $\text{LiNi}_x\text{Co}_{1-x}\text{O}_2$ where $0.5 \leq x \leq 1.0$
3. The battery of claim 1 wherein said positive electrode active material comprises $\text{LiNi}_{0.8}\text{Co}_{0.2}\text{O}_2$.
4. The battery of claim 1 wherein said negative electrode substrate is formed of a material from the group titanium and titanium alloy.
5. The battery of claim 1 wherein said negative electrode substrate is formed of a material from the group nickel and nickel alloy.

6. The battery of claim 1 wherein said negative electrode substrate is comprised of stainless steel.

7. A method of producing a rechargeable lithium battery able to tolerate deep discharge to zero volts without prematurely reducing the battery's capacity, said method comprising:

selecting a positive active material comprising $\text{LiNi}_x\text{Co}_{1-x}\text{O}_2$ (where $0.5 \leq x \leq 1.0$);

coating a layer of said positive active material onto a first metal substrate to form a positive electrode;

selecting a carbon based negative active material;

coating a layer of said negative active material onto a second metal substrate to form a negative electrode;

configuring said positive and negative electrodes to produce at each electrode a Zero Volt Crossing Potential (ZCP) when the voltage between said electrodes is zero; and

configuring each of said electrodes to define a Damage Potential Threshold (DPT) greater than ZCP.

8. The method of claim 7 wherein said step of coating said second metal substrate includes selecting a substrate material from the group consisting of titanium and titanium alloys.

9. The method of claim 7 wherein said step of coating said second metal substrate includes selecting a substrate material from the group consisting of nickel and nickel alloys.

10. The method of claim 7 wherein said step of coating said second metal substrate includes selecting a substrate material of stainless steel.

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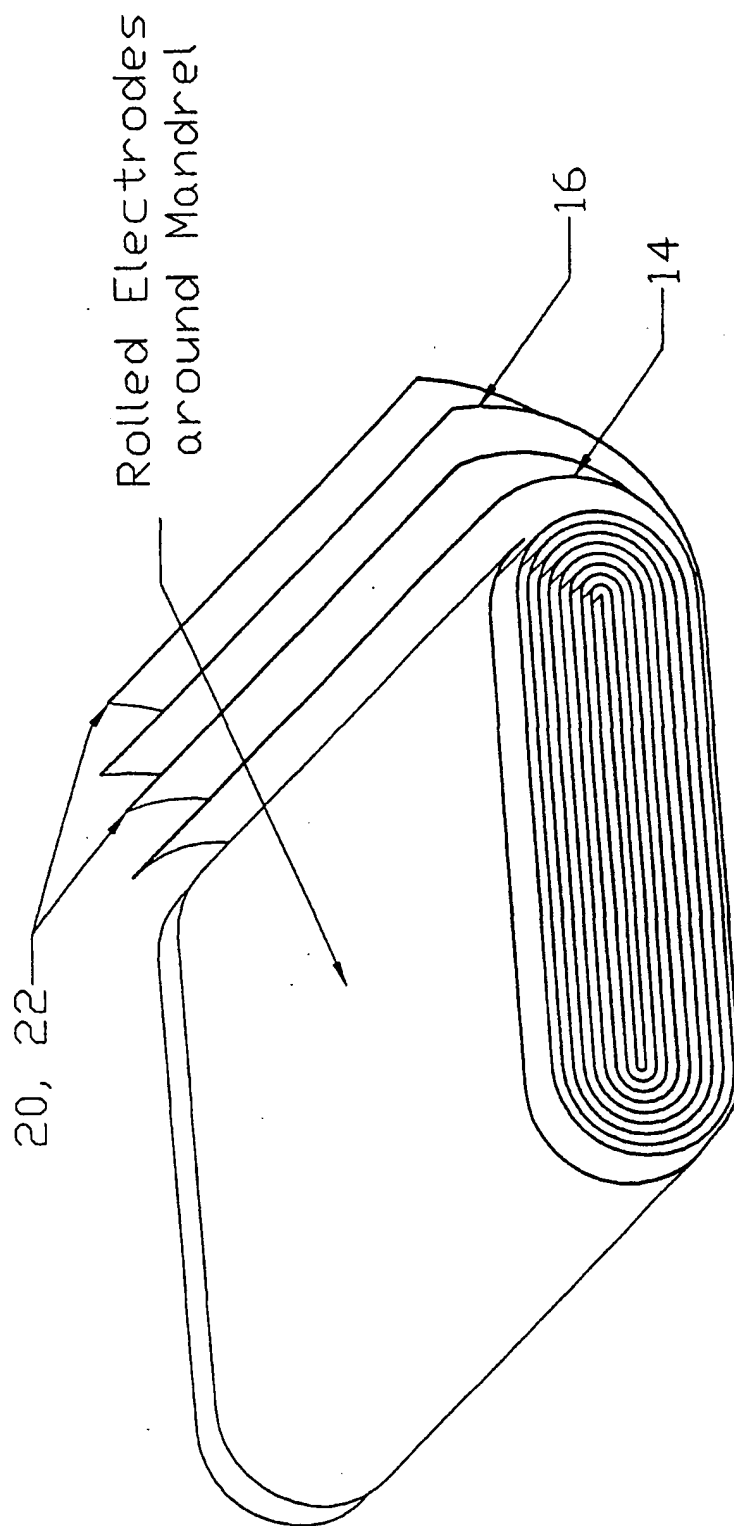


Figure 1A.

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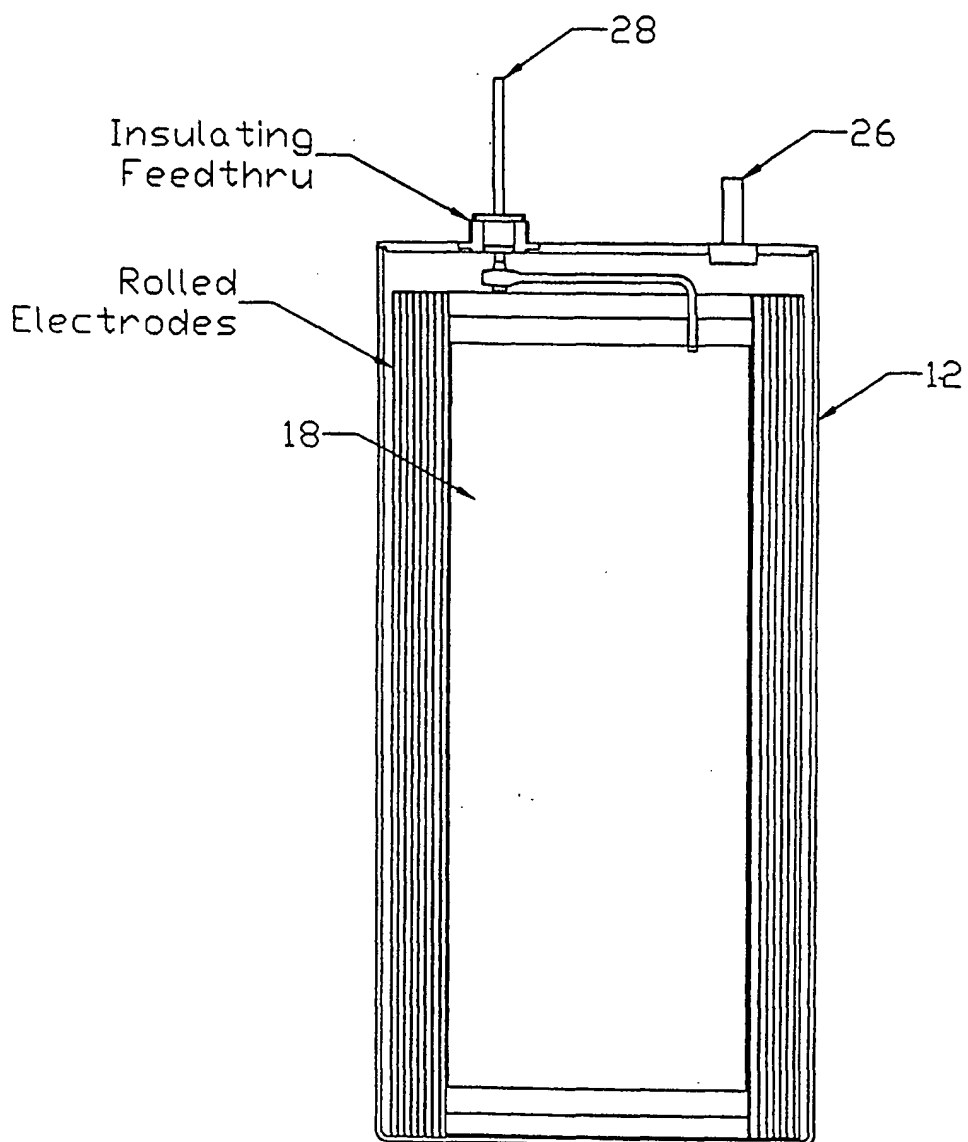


Figure 1B.

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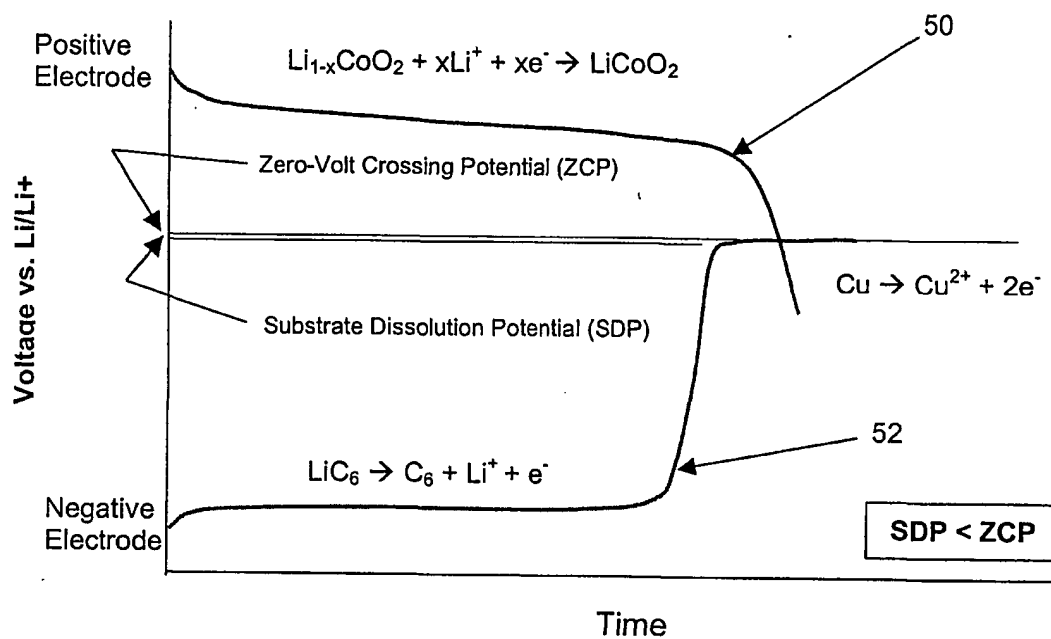


Figure 2.
(Prior Art)

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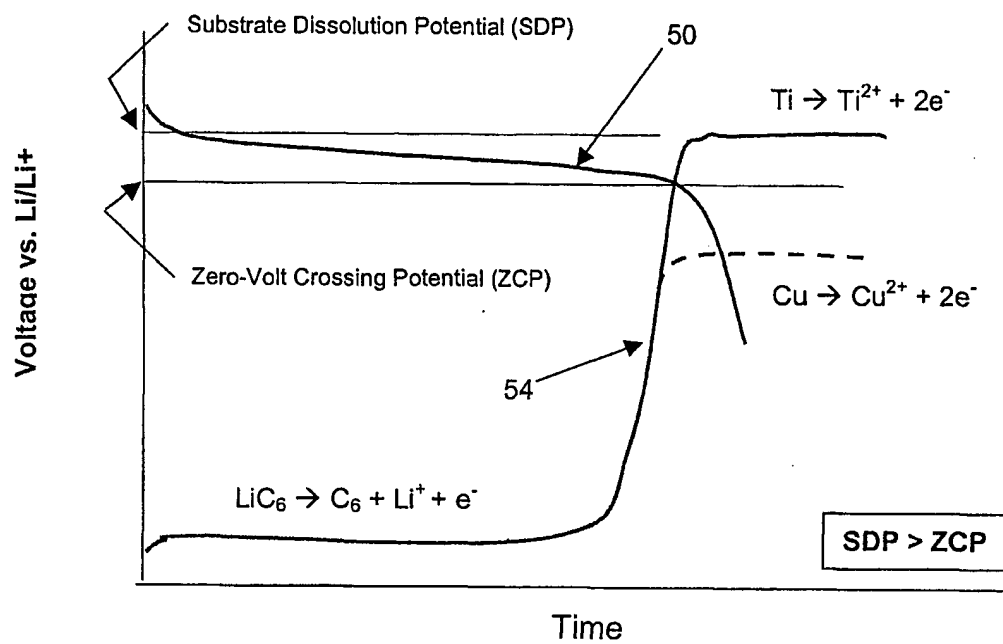


Figure 3.

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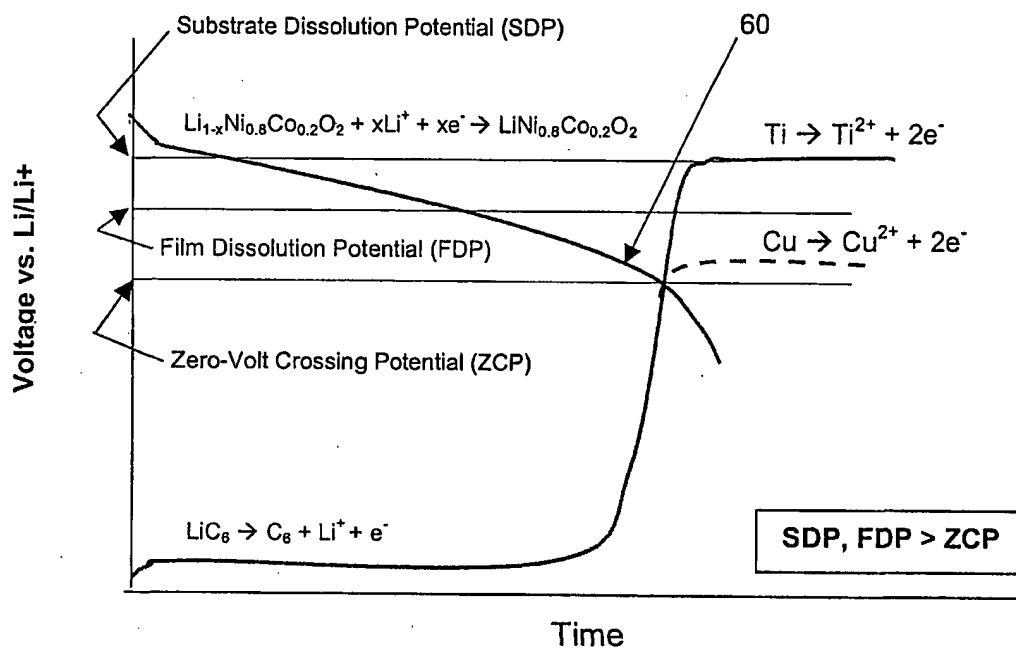


Figure 4.

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	Positive Active Material	Negative Substrate Material	Temperature	Result
(1)	LiCoO_2	Copper	25 °C	Fail; 79.9% Retention
			37 °C	Fail; 76.2% Retention
(2)	LiCoO_2	Titanium	25 °C	Pass; 98.6% Retention
			37 °C	Fail; 73.5% Retention
(3)	$\text{LiNi}_{0.8}\text{Co}_{0.2}\text{O}_2$	Copper	25 °C	---
			37 °C	Pass; 90% Retention
(4)	$\text{LiNi}_{0.8}\text{Co}_{0.2}\text{O}_2$	Titanium	25 °C	---
			37 °C	Pass; 98.8% Retention

Figure 5.

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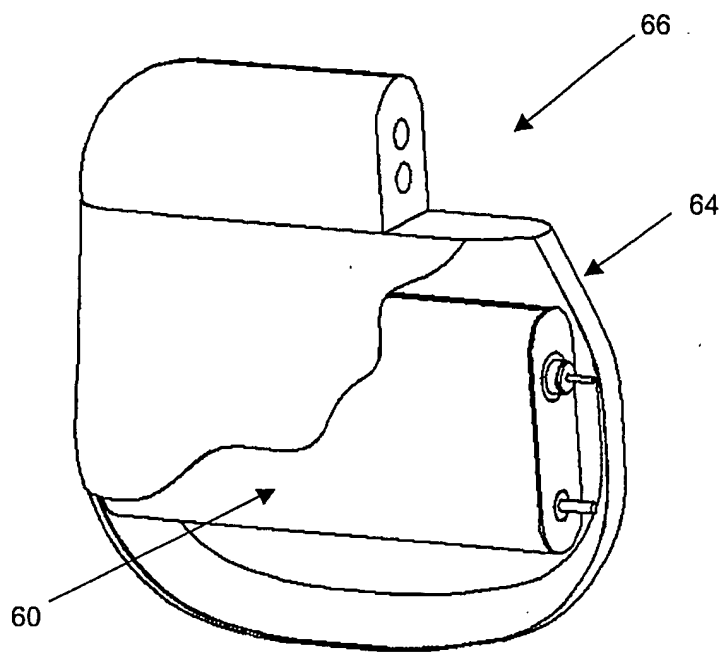


Figure 6.

INTERNATIONAL SEARCH REPORT

International application No.

PCT/US01/43820

A. CLASSIFICATION OF SUBJECT MATTER

IPC(7) : H01M 4/40, 4/52
US CL : 429/223, 231.1, 231.3, 231.8

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 429/223, 231.1, 231.3, 231.8

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
WEST 2.0 (US, EP, JP and Derwent databases)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5,631,100 A (YOSHINO et al.) 20 May 1997, col 3, line 20- col. 4, line 14; col. 8, line 55- col. 9, line 12, claims 14-17.	1, 3-6, 10-11, 13-16 2, 7, 9, 12
Y	US 5,578,398 A (JENKINS et al) 26 November 1996, col. 2, lines 30-40, claims 1-5.	2, 12
Y	US4,092,464 A (DEY et al.) 30 May 1978, col. 5, table 3, claims 1-7.	7-9

☐ Further documents are listed in the continuation of Box C.



See patent family annex.

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Date of the actual completion of the international search

22 March 2002 (22.03.2002)

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06 MAY 2002

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